Metamagnetism of itinerant d-electrons in YCo$_2$: Investigation of metamagnetic transitions in Y(Co, Al)$_2$

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The magnetization of Y(Co$_{1-x}$Al$_x$) intermetallides is studied for strong pulsed magnetic fields $\leqslant 450$ kOe and for $T = 4.2$–300 K. A metamagnetic transition from the paramagnetic to the ferromagnetic state occurs for $x \geqslant 0.075$ at a field whose strength decreases as x increases. The band model for d-electron metamagnetism is used to interpret the results. The metamagnetic transition field 660 kOe and the magnetic moment $1.4\mu_B$/ (formula unit) which characterize the magnetization curve for YCo$_2$ are found by extrapolating to $x = 0$. The results are compared with data for other RCo$_2$ compounds for which the cobalt subsystem has similar properties.

The energy dependence $N(\varepsilon)$ of the density of states and the Fermi energy ε_F determine the behavior of the magnetization curve $M(H)$ for band magnetics. In particular, if $N(\varepsilon)$ for a band paramagnetic decreases near ε_F and has a positive curvature ($N'_{\varepsilon_F} < 0$, $N''_{\varepsilon_F} > 0$, Fig. 1), than the magnetic moment may increase abruptly when an external magnetic field stronger than some critical value H_c is applied. This phenomenon, which was first examined theoretically in Ref. 1, is called metamagnetism of itinerant electrons, or band metamagnetism.

The intermetallide YCo$_2$ (with a cubic crystal structure of the C15 Laves phase type) is a band paramagnetic in which the paramagnetism is enhanced by exchange effects.[2,3] The $N(\varepsilon)$ curve for YCo$_2$ was calculated theoretically in Refs. 4 and 5 by hybridizing the cobalt 3d-electrons to the yttrium 4d-electrons; according to these results, $N(\varepsilon)$ has the qualitative form shown in Fig. 1, which indicates that band metamagnetism may occur in YCo$_2$. No metamagnetic transition has been observed experimentally in YCo$_2$—estimates[4,5] indicate that H_c is $\approx 10^6$ Oe for YCo$_2$, while the magnetization in YCo$_2$ has been studied experimentally[6] only for fields below 380 kOe. Nevertheless, if one postulates the existence of band metamagnetism it becomes possible to explain the observed increase[6] in the susceptibility of YCo$_2$ with H, as well as various magnetic properties of isostructural rare-earth compounds of the form RCo$_2$ (the first-order phase transition from the paramagnetic to the magnetically ordered state, the metamagnetism for T above the ordering temperature, etc.).[7,8]

The band metamagnetism model implies that the critical field H_c of the metamagnetic transition can be reduced by decreasing the density of the d-electrons. This will displace the Fermi surface toward lower energies and increase the state density $N(\varepsilon_F)$ on the Fermi surface (Fig. 1). The susceptibility χ_0 of the d-electrons (neglecting the exchange interaction) will then increase[9]:

$$\chi_0 = \frac{1}{2}(g^2\mu_B^2)N(\varepsilon_F) \tag{1}$$

and so will the exchange-enhanced susceptibility

$$\chi = \chi_0/(1-\alpha\chi_0). \tag{2}$$

Here the exchange coefficient α is related to the exchange integral I in the d-electron system by

$$\alpha = 2I/g^2\mu_B^2. \tag{3}$$

As $N(\varepsilon_F)$ increases, the d-electrons begin to satisfy the Stoner condition

$$IN(\varepsilon_F) \geqslant 1 \tag{4}$$

for band ferromagnetism and the critical field H_c for the metamagnetic transition decreases, as follows from the formula

$$H_c \propto \frac{1}{f}(1-\alpha\chi_0)^{1/2} \tag{5}$$

derived in Ref. 10 for a weak band magnetic. Here the function

$$f = \left[\frac{N''(\varepsilon_F)}{N(\varepsilon_F)} - 3\left(\frac{N'(\varepsilon_F)}{N(\varepsilon_F)}\right)^2\right]^{1/2} \tag{6}$$

may be assumed nearly independent of ε_F for the state density curve shown in Fig. 1.

Experiments have shown that changes in the concentration of d-electrons in RCo$_2$ do indeed alter the magnetic properties considerably.[11,12] In particular, there is a huge increase in the magnetic ordering temperature for RCo$_2$
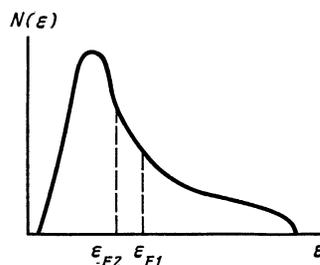

FIG. 1. Rough sketch of the state density curve $N(\varepsilon)$ for YCo$_2$; the values were calculated by hybridizing the 4d yttrium band with the 3d cobalt band; ε_{F1} is the Fermi level in YCo$_2$, ε_{F2} is the shifted Fermi level after substitution of aluminum for cobalt.

when a small fraction of the Co atoms are substituted by Al; we attributed this in Ref. 12 to an increase in $N(\varepsilon_F)$ caused by a drop in the concentration of d-electrons.

In this paper we study how the substitution of cobalt by aluminum alters the magnetic properties of paramagnetic YCo_2. We anticipated that the increase in $N(\varepsilon_F)$ caused by substitution should cause the parameters of the metamagnetic transition to differ substantially from those for YCo_2.

The polycrystalline samples $Y(Co_{1-x}Al_x)_2$ ($0 \leqslant x \leqslant 0.2$, $\Delta x = 0.025$) were fused from the starting materials in an arc furnace (cold floor, argon atmosphere, noneroding tungsten electrode). The samples were annealed for 200 h at 850 °C inside a vacuum chamber which was continuously pumped down to 10^{-4} torr. Less than 0.1 wt.% of the material was wasted during synthesis.

It was noted in Ref. 3 that in order to synthesize monophase YCo_2 samples, the starting mixture should contain a small excess of yttrium; the excess was 4–5 wt.% in our experiments. We verified by x-ray and metallographic analysis that only a single phase was present (these measurements were accurate to 1–2%). All samples shown to contain impurity phases were discarded. We were able to synthesize monophase samples only for $x \leqslant 0.15$; for larger x the samples contained a mixture of the C15 phase with the hexagonal C14 phase. [The C14 phase is known[13] to be present in $Y(Co_{1-x}Al_x)_2$ for $0.2 \leqslant x \leqslant 0.8$.]

We used the induction method to measure the magnetization for $4.2 \leqslant T \leqslant 300$ K in pulsed magnetic fields $\leqslant 320$ kOe, while a vibration magnetometer was employed for dc fields $\leqslant 60$ kOe (fields $\leqslant 450$ kOe were used in the measurements at $T = 4.2$ K in Refs. 14, 15).

Figure 2 shows the field dependence $M(H)$ for $Y(Co_{1-x}Al_x)_2$ at $T = 4.2$ K. We see that M depends almost linearly on H for YCo_2 and $Y(Co_{0.95}Al_{0.05})_2$—these compositions were paramagnetic for the fields investigated. For higher aluminum contents x there was a range of fields for which M increased faster than linearly, and a metamagnetic transition occurred for these compositions. The transition shifted toward lower fields as x increased. It is also noteworthy that the transition did not occur abruptly in our samples but extended over a range of fields that became narrower as x decreased. The metamagnetic transition for $x = 0.15$ was so broad that the increase in M was almost smooth. Figure 2 shows that $M(H)$ for strong H dropped as x increased, and calculations show that this decrease cannot
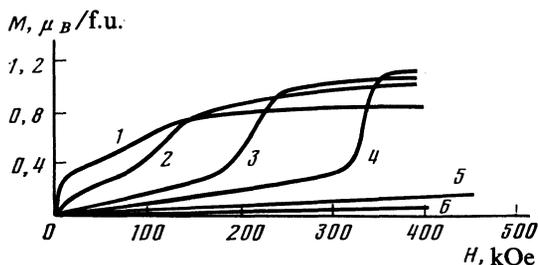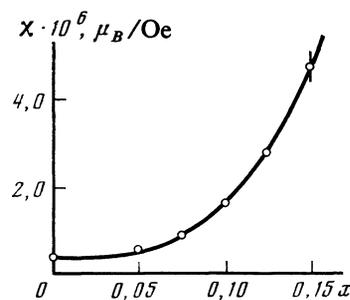

FIG. 3. Paramagnetic susceptibility of $Y(Co_{1-x}Al_x)_2$ as a function of x; $T = 4.2$ K.

be due simply to the decreased Co content—the magnetic moment calculated per Co atom drops as x increases.

Our data show that the susceptibility in relatively weak fields (below H_c for the metamagnetic transition) rises abruptly as x increases (Fig. 3). There is also a slight spontaneous magnetization (~ 0.1–$0.2\,\mu_B$) in samples with large x. This magnetization is apparently a property of the matrix and is not caused by the impurity. This follows from our x-ray studies of the crystal lattice parameters for the fundamental C15 phase in $Y(Co_{0.85}Al_{0.15})_2$ for $T = 5$–300 K. We found that for $T \lesssim 50$ K the temperature dependence of the lattice parameter deviated from the Debye curve typical for paramagnetic YCo_2. This deviation may be attributed to magnetic ordering of the d-electron subsystem, which gives rise to anomalous bulk magnetic behavior (such behavior, although much more pronounced, is observed[16] in the magnetically ordered RCo_2 compounds).

The metamagnetic transition became blurred as T increased and in all cases $M(H)$ was nearly linear for $T \gtrsim 50$–70 K. As an example Fig. 4 plots $M(H)$ for $Y(Co_{0.90}Al_{0.10})_2$ recorded for several temperatures.

Our data thus confirm that YCo_2 is a band metamagnetic, and that changes in the d-electron concentration alter the metamagnetic transition field as predicted above. The experimental findings can be accounted for qualitatively by the rigid band model, according to which the added aluminum leaves the state density $N(\varepsilon)$ unchanged and simply decreases the population of the d-band. Because of the behavior of the energy dependence $N(\varepsilon)$ (cf. Fig. 1), this causes $N(\varepsilon_F)$ to increase.
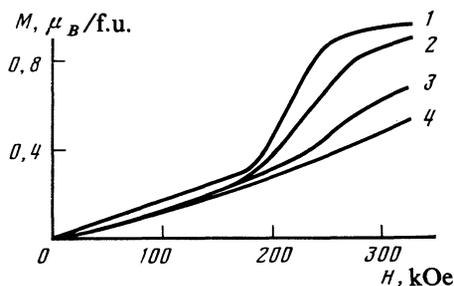

FIG. 2. Magnetization curves for $Y(Co_{1-x}Al_x)_2$ at 4.2 K: 1) $x = 0.15$; 2) 0.125; 3) 0.10; 4) 0.075; 5) 0.05; 6) 0.

FIG. 4. Magnetization curves for $Y(Co_{0.90}Al_{0.10})_2$ for $T = 4.2$ (1), 40 (2), 60 (3), and 80 K (4).

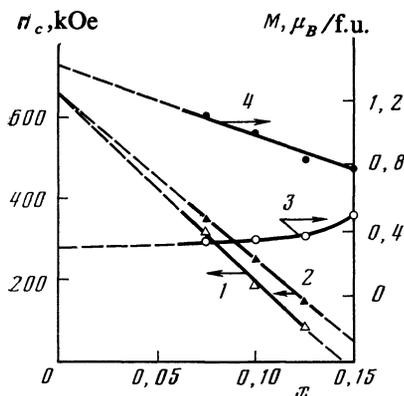154   Sov. Phys. JETP **62** (1), July 1985

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FIG. 5. Concentration dependence of the fields H_{c1} ($\triangle$) and H_{c2} ($\blacktriangle$) at which the metamagnetic transition begins and ends, and the magnetic moments at $H = H_{c1}$ and H_{c2} ($\bigcirc$ and $\bullet$, respectively); $T = 4.2$ K.

the magnetic moments M_{C01} and M_{C02} at these fields—vary linearly with x for $x < 0.15$ (Fig. 5). We can therefore extrapolate these values linearly to $x = 0$ to get $H_c \approx 660 \pm 50$ kOe for pure YCo_2, for which the metamagnetic transition should be abrupt; the magnetic moment of cobalt at $H = H_c$ is $M_{C01} = (0.35 \pm 0.1)\mu_B$/f.u., and $M_{C02} = (1.4 \pm 0.1)$ μ_B/f.u. By comparison, the theoretical estimates in Refs. 4 and 5 give $H_c = 10^6$ Oe, $M_{Co2} \approx 2.0\mu_B$/f.u. and $H_c = 3.5 \cdot 10^6$ Oe, $M_{Co2} = 1.8$ μ_B/f.u., respectively; moreover, the data in Ref. 7 for RCo_2 imply that $M_{C02} \sim 2\mu_B$/f.u. In view of the various approximations made in these calculations, the agreement may be regarded as satisfactory.

We have thus experimentally detected band metamagnetism in $Y(Co_{1-x}Al_x)_2$, studied how the metamagnetic properties depend on x, and determined the properties of the magnetization curve for YCo_2 for fields $H \approx H_c$. The results are comparable to those found for other compounds RCo_2 in which the cobalt subsystem has similar properties.

However, such a simple model is very crude at best. For example, it ignores the fact that substitution alters the *s*-electron concentration and thereby changes the contribution from *s-d* exchange. In addition, the unit cell volume increases with x, so that the overlap between the *d*-electron wave functions is decreased. These factors can also alter the exchange integral I and the state density $N(\varepsilon_F)$ that determine when system (4) becomes magnetically ordered. However, the latter effect is apparently small. This follows by comparing our data on $\chi(x)$ for $Y(Co_{1-x}Al_x)_2$ with the results in Ref. 17, where the effects of isotropic compression on χ were studied for YCo_2. Indeed, the bulk effect $d \ln\chi / d(\ln V) = 14 \pm 2$ found there is an order of magnitude less than the observed increase in χ for $Y(Co_{1-x}Al_x)_2$.

Finally, we note that because the aluminum atoms are randomly distributed in the lattice, structural inhomogeneities are present in the mixed compounds $Y(Co_{1-x}Al_x)_2$ and may form clusters that broaden the metamagnetic transition. The spontaneous magnetization of $Y(Co_{1-x}Al_x)_2$ for large x is in all probability due to the presence of Al-rich clusters in which the cobalt atoms are already magnetically aligned. However, the spontaneous magnetization can also be described by the band metamagnetism model[18] without any consideration of structural inhomogeneities.

Analysis of the experimental data shows that the magnetic properties of $Y(Co_{1-x}Al_x)_2$—the fields H_{c1} and H_{c2} at which the metamagnetic transition begins and ends, and

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